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WORK OF ACADEMICIAN S. V. LEBEDEV IN THE FIELD OF SYNTHETIC RUBBER

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Lebedev must be regarded as a pioneer in the field of synthetic rubber in the sense that he first introduced and successfully applied a workable industrial process for the synthesis of butadiene rubber. On the basis of the insufficient knowledge of the subject which was available in 1908, the year in which Lebedev started his work, he quickly realized that attempts aiming at an exact duplication of the natural product were both wasteful and impracticable. During the period 1908 - 1912 Lebedev thoroughly investigated the polymerization of 16 hydrocarbons of the butadiene series and of several derivatives of allene. His results were published as a comprehensive monograph in 1913. In the course of this work, Lebedev determined, the structure of the products by decomposing them with ozone (Harries' method) and carried out a far-reaching study of the kinetics of polymerization. He postulated a mechanism of chain growth which approaches quite closely the commonly accepted modern theory. He first realized the importance of active centers (or active complexes) and assumed that polarization of the molecules of the monomer play an important part in polymerizations. The following relationships were established by Lebedev:

"As temperature increases, the ratio of the quantity of the dimer to that of the polymer is shifted in favor of the dimer.

"1. In a series of isomers having a conjugated system, the rate of polymerization increases as the substituent is moved from the end atoms to the middle of the molecule.

"2. Ring formation by a conjugated chain increases the rate of polymerization.

"3. In a homologous series, an increase of the mass of a substituent in the middle portion of a conjugated system increases the rate of polymerization, while an increase of the mass of a substituent attached to an end atom reduces that rate, assuming that heating is conducted at comparable temperatures."

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These relationships were formulated and published by Lebedev in 1913 or shortly thereafter in a special Russian monograph and in a paper appearing in the Journal of the Russian Chemical Society. Lebedev's studies permitted a rational approach to the selection of a suitable starting material for the synthesis of rubber. His results could be actually applied in 1925, when the Soviet government announced a contest for a satisfactory industrial method of rubber synthesis. While Germany produced synthetic dimethyl butadiene rubber during World War I (I. L. Kondakov found in 1899 - 1901 that polymerization of dimethyl dutadiene led to an elastomer), the product was generally regarded as unsatisfactory and too expensive. In his work aiming at an industrial synthesis, Lebedev proceeded independently and carried out further important research during 1926 - 1928. On the basis of Lebedev's prior experience, butadiene was chosen as the starting material. Two methods for preparing butadiene were open: synthesis from ethyl alcohol and preparation from petroleum raw materials.

The first method led rapidly to a satisfactory result. The known process discovered by Ostromyslenskiy (dehydrogenation of ethyl alcohol and subsequent dehydration, i.e., two stages) gave a yield of no more than 5 - 6 percent of the first product. Lebedev improved the procedure and increased the yield by carrying out dehydration of the formed aldehyde simultaneously with dehydrogenation in one stage over a mixed catalyst. Later, a great number of catalysts were tested in this connection and a yield approaching the theoretical was achieved. In solving the complex problem of polymerization, Lebedev developed a satisfactory procedure using sodium and alkali metals in general, although this method had been discarded by Harries and Ostromyslenskiy. Having in mind the inadequate yields obtained by the Germans, who polymerized by heating (thus obtaining too much of the dimer), Lebedev realized the advantage of carrying out the polymerization at a low temperature in the presence of alkali metal. Rapidity of conversion and the uncomplicated equipment required were other advantages of the method selected and developed by Lebedev. Lebedev was also aware of the shortcomings of the method, but the choice he made was the only correct one at that time. On the basis of results presented to the commission on 1 January 1928, Lebedev was declared the winner of the contest and the government assigned funds for future work. A special laboratory of synthetic rubber was founded and attached to the petroleum laboratory at the University of Leningrad.

During 1928 - 1929 the necessary data for construction of a pilot plant was obtained in that laboratory. In 1927 - 1928, Lebedev found that the strength of synthetic rubber could be increased by adding fillers, and that the effect of fillers in that respect was much greater than with natural rubber. The pilot plant was started in 1930, and the first full-scale industrial plant producing butadiene rubber began operations in 1932.

In 1936 - 1938, a synthetic rubber industry based on butadiene was created in Germany. In 1941 - 1943, production of butadiene synthetic rubber was introduced in the US.

A further achievements which must be credited to Lebedev is polymerization of ethylene hydrocarbons by means of silicates, a process which forms the basis of the synthesis of isobutylene elastomers. Studies by Lebedev and collaborators demonstrated that depolymerization of isobutylene polymers predominates at high temperatures. The polymerization was carried out at temperatures as low as minus 125 degrees, and polymers having an average molecular weight of 8,000 were obtained. In 1929 - 1930, Lebedev's collaborators began work on the synthesis of butadiene starting with butylenes (dehydrogenation of butylenes). Death interrupted Lebedev's activity in 1934.

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